/N-47
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1.Characterization of Aircraft Produced Soot and Contrails near the Tropopause
2. Summary of Research
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4. 6/1/94 to 12/31/97
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6. NAG 2-923

I. **Introduction.**

Participation in the SUCCESS project primarily involved development and deployment of specific instruments for characterizing jet aircraft exhaust emissions as particulates and their subsequent evolution as contrail particles, either liquid or solid, as cirrus. Observations can be conveniently considered in two categories - close or distant from the aircraft. Thus close to the aircraft the exhaust is mixing through the engine turbulence with a much drier and colder environment and developing water/ ice supersaturation along the trail depending on circumstances (near field), whereas distant from the aircraft (far field) the exhaust has cooled essentially to ambient temperature, the turbulence has decayed and any particle growth or evaporation is controlled by the prevailing ambient conditions. Intermediate between these two regions the main aircraft vortices form (one on each side of the aircraft) which tend to inhibit mixing under some conditions, a region extending from a few aircraft lengths to sometimes a hundred times this distance. Our approach to the problem lay in experience gained in characterizing the smoke from hydrocarbon combustion in terms of its cloud forming properties and its potential influence on the radiation properties of the smoke and subsequent cloud from the viewpoint of reduction (absorbtion and scattering) of solar radiation flux leading to significant global cooling (Hudson et al 1991; Hallett and Hudson 1991). Engine exhaust contains a much smaller proportion of the fuel carbon than is sometimes present in ordinary combustion (<0.01% compared with 10%) and influences condensation in quite different ways, to be characterized by the Cloud Condensation Nucleus, CCN - supersaturation spectrum. The transition to ice is to be related to the dilution of solution droplets to freeze by homogeneous nucleation at temperatures somewhat below -40C (Pueschel et al 1998). The subsequent growth of ice particles depends critically on temperature, supersaturation and to some extent pressure, as is demonstrated in an NSF funded project being carried out in parallel with the work reported here. As will be discussed below, nucleation processes themselves and also exhaust impurities also influence the growth of ice particles and may control some aspects of growth of ice in contrails. Instrumentation was designed to give insight into these questions and to be flown on the NASA DC-8 as a platform. In addition a modest program was undertaken to investigate the properties of laboratory produced smoke produced under controlled conditions from the viewpoint of forming both CCN and CN. The composition of the smoke could inferred from a thermal characterization technique; larger particles were captured by formvar replicator for detailed analysis; ice particles were captured and evaporated in flight on a new instrument, the cloudscope, to give their mass, density and impurity content.

II Scientific Questions.

We examine the scientific questions which we perceived as driving our participation in the project. The fundamental mechanism to be probed is whether and how particles emitted from engine exhausts lead to contrail formation under different conditions in the atmosphere. A further question is whether such particles can (prior to removal by various scavenging process) be recycled to form much more extensive upper tropospheric cloud as increased area coverage or thicker (greater optical depth) cloud. Such events could lead to a change in the earth's radiation balance resulting in changes in global temperature and precipitation distribution. A parallel question is related to fuel composition and whether impurity removal or additives could change the situation - or whether indeed modest changes in engine design could be undertaken. Such considerations lead to questions on particle nucleation, particle growth and evaporation from a trajectory from engine exhaust to how ultimately the particles are scavenged out by precipitation. Specific instrumentation has been designed, built and used to answer aspects of these questions. It is to be pointed out here that particles not only interfere with radiation but also act as substrates for catalyzing chemical reactions so that our findings may have relevance to ozone chemistry particularly at low temperature (Carslaw et al 1998).

III Results.

Summarized below are the general results that follow from the listed publications in section V and publications in preparation:

* Aircraft measurements of aircraft exhaust CCN (0.2 -1%) and CCN/CN ratio at contrail levels showed substantially higher values than had been measured at lower levels in the atmosphere. A possible explanation lies in the presence of very clean air , as a product of previously existing cirrus, leading to an effective scavenging of CCN as they dilute, nucleate ice and are removed by ice precipitation to somewhat lower levels where ice evaporation occurs (Figure 1). Thus ice precipitation and the presence of very high values of CN is an observation consistent with fresh nucleation in the *absence* of CCN to provide a basis for chemical reactions (Hudson and Xie, 1998).

* These results together suggest that although the high and low sulfur fuel may give rise to exhaust with high and low vales of CCN, there may be somewhat of a feedback process whereby ultimately natural ice processes may give a dearth of CCN which in turn leads to high CN nucleation and more CCN following aggregation. The net result in either event would be some increase in cirrus cloud presence (area) and/or optical depth with the presence of more sulfur in the exhaust. These considerations show that we need to obtain better insight into the overall aerosol particle economy of clouds and their environments at these levels. (In preparation).

* Measurements of CCN were made of smoke produced from high and low sulfur fuel in a laboratory test burn. In both cases CCN for high sulfur fuel was substantially (X 10) above that for low sulfur fuel suggesting an increased hygroscpic component of the particles; heating the high sulfur sample to 180C reduced the CCN concentration in the smoke by about X 20, whereas heating had a minimal effect on smoke from low sulfur smoke, (Hallett et al 1997).

* Particles collected on replica in an evaporating contrail were small (10 microns) , had excessive trigonal symmetry, (suggesting a defect/impurity effect) and concentrations up to 1 per ml, (figures 2, 3 ,4). The *variability* appeared greater for flight segments from low sulfur fuel. Such particles were not collected on the cloudscope, suggesting a density of less than 0.2 and a low collection efficiency (Meyers & Hallett, 1998; Meyers, Arnott and Hallett in preparation). This low density is consistent with rapid evaporation rate of similar larger ice particles observed on the cloudscope under other circumstances. It appears that a higher proportion of crystals with trigonal symmetry was associated with higher sulfur fuel burn.

* Cloudscope collection of droplets in a wave cloud over Boulder showed a droplet concentration of somewhat less than 10 per ml of size some 15 microns. These numbers do not differ substantially from the concentration of ice particles (Jensen et al 1998). Comparison with CCN measurements suggested that only a very small fraction of nuclei were activated requiring a supersaturation of below 0.1 per cent over water. As these droplets evaporated, no visible residue remained, consistent with the solid residue being well below one micron in diameter, the limit of resolution, irrespective of its composition. (Hudson and Hallett in preparation).

* Accretion and evaporation of ice particles on the cloudscope at low temperature (<-55C) showed, on occasion, the apparent melting of the ice as evaporation progressed. The observation is consistent with the presence of a H_2SO_4 residue and dissolution of the ice as the solution concentration increases. This we consider as an important observation, but is very tedious in analysis and is still under study.

IV **Instrumentation**.

Existing instruments were adapted for installation on the DC-8 and modified for the specific task of looking at the nature of particles in aircraft contrails. Instrument location and details of the missions are shown in table 1.

A. The cloudscope.

The cloudscope was first constructed as an operating system in 1995 (Arnott et al 1995) and has been developed into a highly reliable instrument for aircraft deployment. The operating principle is collection of particles near the stagnation point on a forward facing optical flat, video recorded from behind with a resolution of about 2 microns. Collection falls for sizes of particle below some 10 microns such that 2 micron diameter particles are not collected (Figure 5). The rate of change of particle area following evaporation in the compressed air is to be related to particle density (Hallett et al 1998). The instrument fits a PMS pod and is mounted under the wing tip of the DC-8. Modifications were made for the SUCCESS mission to improve illumination of the particles and the ease of mounting in the pod so that instruments could be exchanged as required. Data analysis (Arnott et al 1998; Hallett et al 1998) uses a frame grabbing technique to capture the video images and estimates their area differences during evaporation. The spatial concentration is estimated from a knowledge of particle density, airspeed and the dimension of the collecting housing. Shape (habit) is estimated directly from the image. It is usually possible to obtain some estimate of hygroscopicity of such particles by observing their behavior on the cloudscope under conditions of increasing relative humidity.

* The Replicator.

The principle of operation is to collect particles in a thin film of solution of formvar on a moving 70 m length of 16mm film. The solution dries by evaporation of the solvent to leave a cast of the ice or water particle, to be examined by microscopy in the laboratory (Hallett 1976; Arnott et al 1998). The version used in the DC-8 fits a PMS pod and was, for this deployment, ventilated to give a particle collection efficiency down to about 1 micron. In practice this resolution was a few microns because of impurity particles in the solution. The film speed could be changed from 1/2 or 1 cm per second as required to give a sample time of up to 3 hrs. Modifications were made for this mission to improve the speed of deployment prior to each flight and in the viewing necessary to give better quality control during flight. Reduced data are in the form of size, habit and phase distributions, together with detailed images by optical and scanning electron microscopy.

C. The CCN Spectrometer.

The principle of operation is to use a vertical water vapor thermal diffusion chamber to produce a gradient of supersaturation through which outside air passes in a sheath flow allowing activation of aerosol which grow and are sensed optically as they leave the chamber (Hudson 1989). The instrument is mounted in the cabin and samples air from a by pass flow as required. The aerosol either passes to the chamber directly or through a temperature processor operating up to a selected temperature of 500C. The chamber operates at ambient pressure and concentration reduced to a standard pressure for inter comparison. A supersaturation spectrum requires about 5 seconds and is plotted as a differential concentration - supersaturation; the spectrum may be extrapolated to higher/ lower supersaturation as required thin limits of accuracy given by the spectrum variability (Figure 6). CCN results were compared with CN data obtained by P.Demott/ D.Rogers of CSU.

V Aircraft Instrument Comparison.

During the course of this work and previous related projects, it has become necessary to collect data from different instruments measuring different quantities which are not co-located (Figure 7). Instruments are mounted on each wing tip and also at various locations on the fuselage and such data need to be synthesized in order to compute derived quantities, such as aerosol flux as a product of aerosol number concentration (or area or mass) and vertical velocity for example. A similar synthesis is involved in combining two instruments measuring different size ranges in a particle spectrum - as small ice particles tens of microns and larger ice particles many hundreds of microns. In as far as there is an approximate exponential relationship through an empirical gamma distribution or a theoretical relation ship through a Weibull distribution (Liu and Hallett 1998), there tends to be a smaller concentration of larger particles. It follows that at some point for any particle counting instrument a choice has to be made in assessing particles from the viewpoint of Poisson statistics as the sample volume (or time) gives a small number of particles in the sample interval, it being remembered that a concentration N per sampled volume in some time interval t has a variance in a uniform field of $N^{1/2}$. Thus assuming that instruments on each wing tip 'see' air with the same uniform property, comparison, as a test for uniformity can only be achieved within these limits. In a true uniform field with random distribution the sample time can, in principle, be extended indefinitely; in a finite field limitations always exist. A casual window view of the spatial distribution of particles in a contrail sampling mission dramatically brings home this point - sometimes only one wing tip lies in the contrail. And this is not all - when the contrail itself is wrapped into the aircraft wing tip vortex, spatial variability exists as particle free contrail air is interleaved with particle containing contrail air. Inhogeneities down to a scale of meters and below readily exist. Differing airflow at the site of collection resulting from aircraft flight changes present an additional difficulty.

A conclusion from these considerations is that while data from a single instrument yields useful results over its (limited) dynamic range, combination of a spectrum from two instruments differently located has to be treated with caution. The same argument holds for comparison with integrated water, as with an ice mass content instrument as a CVI. These thoughts are common to all aircraft measurements and are dominating attempts at instrument comparison in numerous cases in the SUCCESS mission. The problem is particularly worrying for contrail studies as the intrinsic spatial scales are necessarily short. For larger scales (as natural cirrus) intrinsic variability for two instruments need to be derived and any comparison made on this basis. A paper is in preparation on this topic.

VI Related Project:

In parallel with this NASA study, a NSF funded laboratory study has been undertaken of the habit of ice crystals growing from the vapor under cirrus conditions. Specifically a water vapor thermal diffusion chamber has been constructed which enables temperature, supersaturation over ice and air pressure to be controlled independently. The temperature range covers -30C to below -70C, supersaturation from a few% to over 200% over ice, and air pressure from ambient 850 to 1 mb. Ice crystals are grown on a narrow vertical filament and the growth rates and habits video recorded. Such habits can be mapped and related to cirrus conditions as in Figure 8. The effective supersaturation is estimated from the ventilation coefficient of a crystal falling at terminal velocity. (Bailey and Hallett, 1998). Future studies will involve addition of known impurities to the chamber atmosphere to provide a series of maps similar to Figure 8.

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*Hallett, J., J.G. Hudson, F. Rogers, E. Teets, S.S. Yum and Y. Xie, 1997, The Influence of Natural and Aircraft Exhaust on Aerosol and Cirrus Formation. Third Conference on Atmospheric Chemistry, AMS, February 1997, Long Beach CA pp.45 - 53.

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*Jensen, E.J., O.B. Toon, A. Tabazadeh, G.W. Sachse, B.E. Anderson, K.R. Chan, C.W. Twohy, B. Gandrud, S.M. Aulenbach, A. Heymsfield, J. Hallett, and B. Gary, 1998. Paper No. 98GL00299, Ice nucleation processes in upper tropospheric wave-clouds observed during SUCCESS. *Geophys. Res. Lett.*, **25**, pp.1363-1366.

*Poellot, M.R., W.P. Arnott and J. Hallett, 1998, In Situ Observations of Contrail Microphysics and Implications for their Radiative Budget. J.G.R., In Press.

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†Arnott, W.P., T.J. Ulrich, R.Cole and J.Hallett: 1998; Summary of in-situ observations of Cirrus Microphysics During FIRE II: Instrumental Response. Conference on Cloud Physics, Everett, WA, August 1998.

†Bailey, M. and J. Hallett, 1998; Laboratory investigation ice growth from the vapor under cirrus conditions between -30C and -70C, Conference on Cloud Physics, Everett, WA, August 1998, pp.434-437.

*Meyers, M. and J. Hallett 1998 Ice crystal Morphology in Aircraft contrails and Cirrus, Conference on Cloud Physics, Everett, WA, August 1998, pp.17-20.

†Schmitt,C., W.P. Arnott and J. Hallett,1998; Infrared Extinction and Emission by Laboratory Ice Clouds: Simulation of Cold Cirrus and Contrail Radiative Properties. Conference on Cloud Physics, Everett, WA, August 1998.

Presentations to be made at the CIRRUS Conference in Baltimore, October 1998:

†Shape and Fall of Cirrus Particles.
(Invited paper) John Hallett and Pat Arnott (DRI).

†New Insight for in situ quantification of cirrus
W. Patrick Arnott and John Hallett (DRI) and Michael R. Poellot (UND)

Papers in Preparation:

†Laboratory study of cirrus growth habit related to environmental parameters.
Matthew Bailey and John Hallett

*Kinne, S., J. Hallett, et al
Intercomparison of aircraft measurements by instruments mounted at different locations.

*Hallett,J., J.G.Hudson, F.Rogers, E.Teets, S.S. Yum and Y. Xie, The Influence of Natural and Aircraft Exhaust on Aerosol and Cirrus Formation.

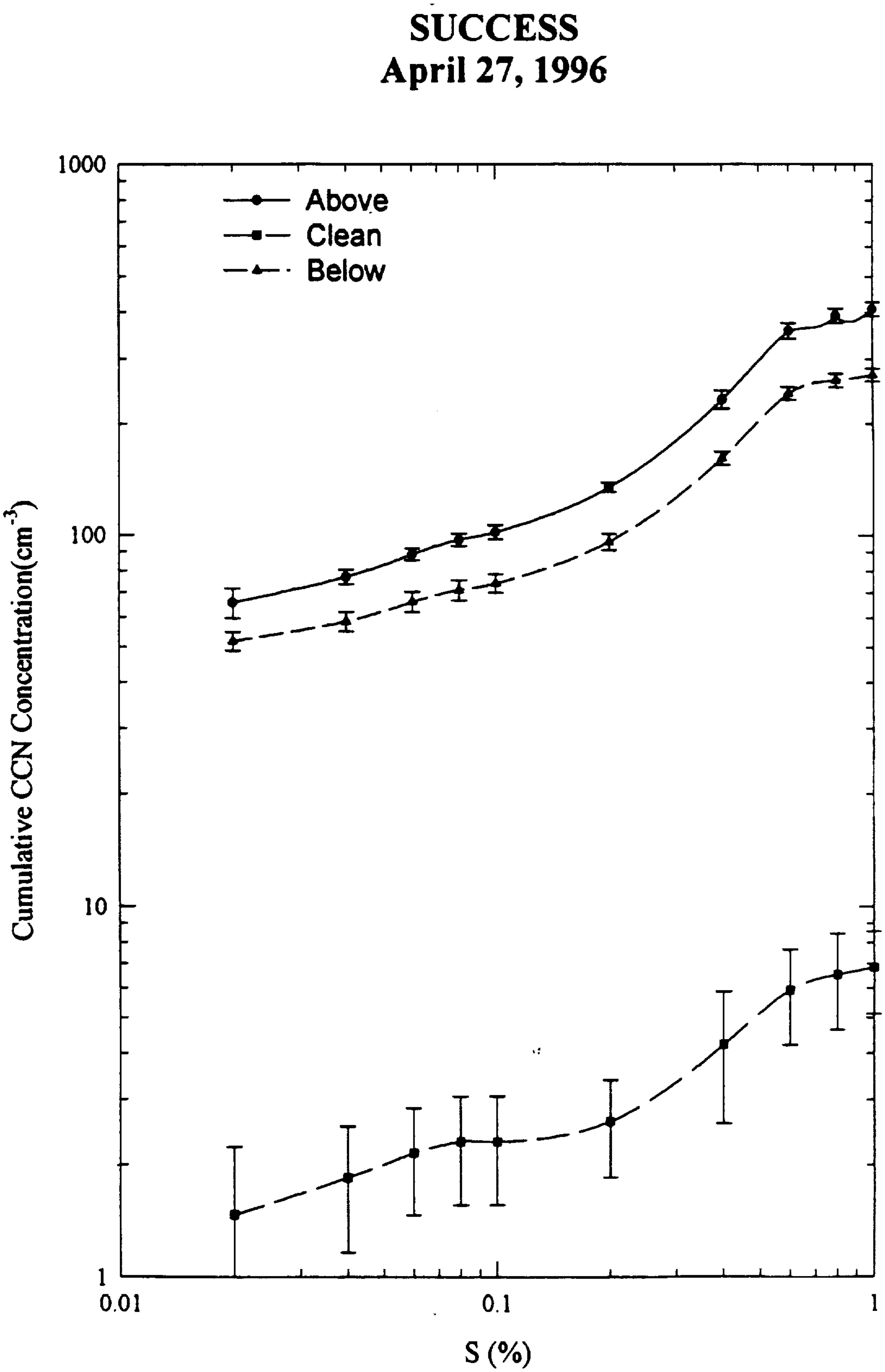
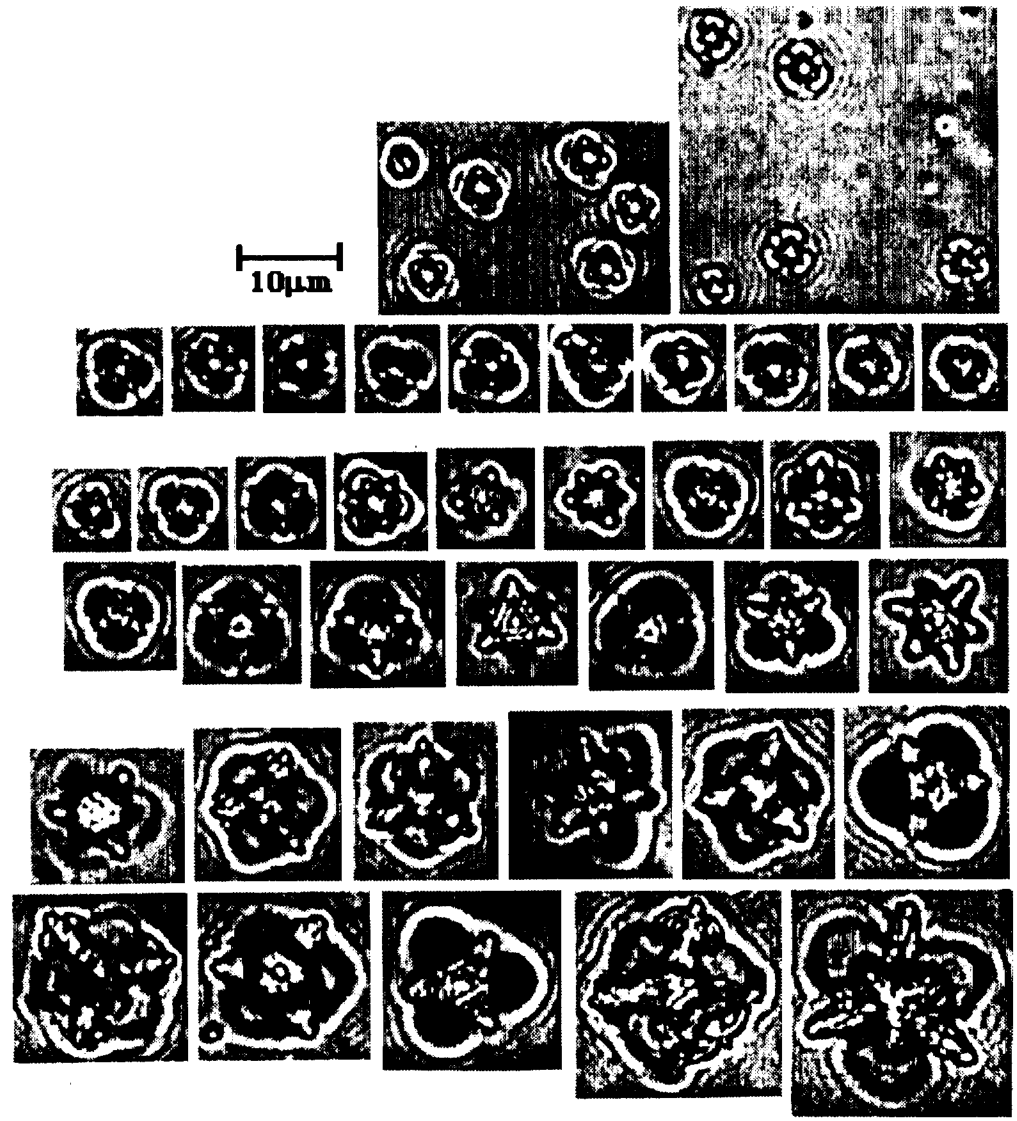


Figure 1. CCN spectra at contrail levels showing high variability in and out of old contrail regions.

Symmetrical Spiny crystals are most commonly found in high sulfur contrail

- Six pointed
- Three dimensional
- Fanning points

- High concentration
- Generally < 10μm
- High sulfur 32.5%
- Low sulfur 7.3%



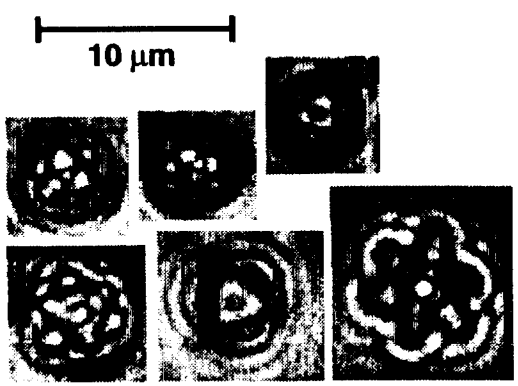
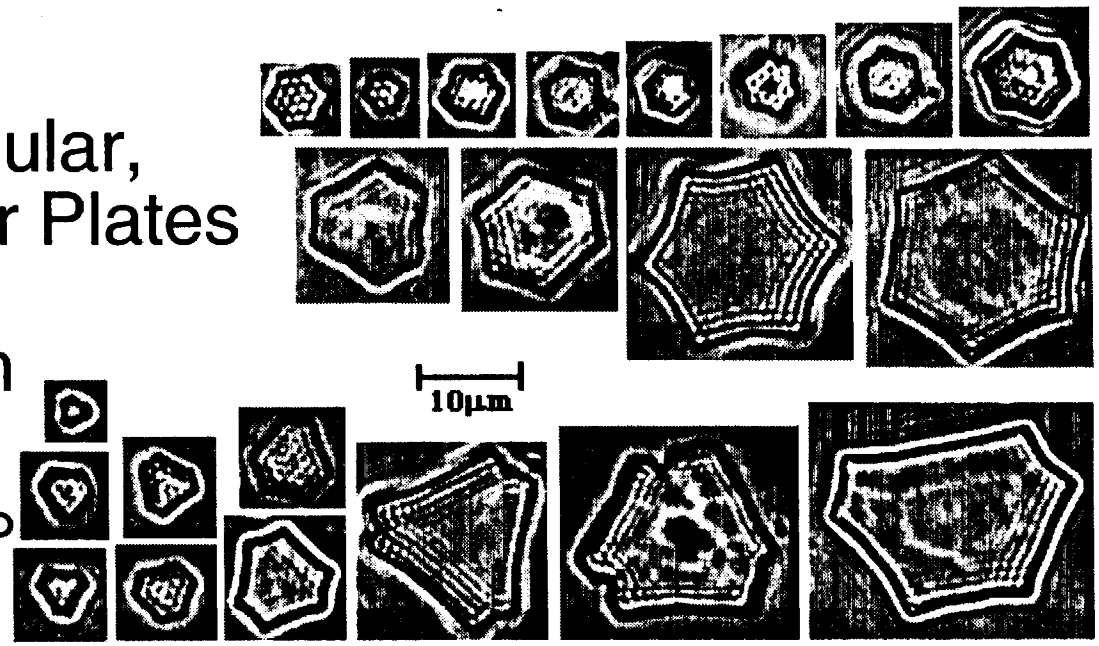


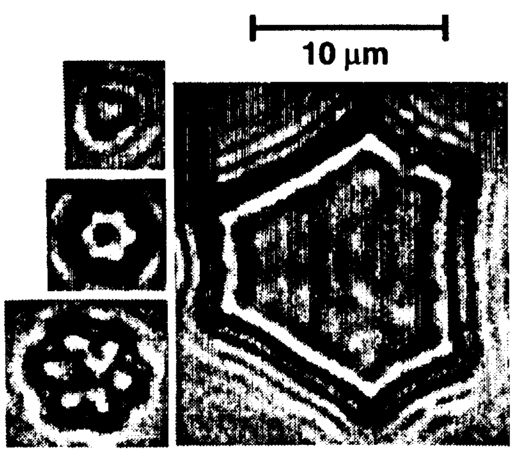
Figure 2. Replica of ice crystals from an evaporating contrail showing three dimensional trigonal structure.

Plate-like crystals dominate the concentration

- Hexagonal, Triangular, Scalene, and Sector Plates

- High concentration
- Generally < 10μm
- High Sulfur 65.0%
- Low Sulfur 89.5%





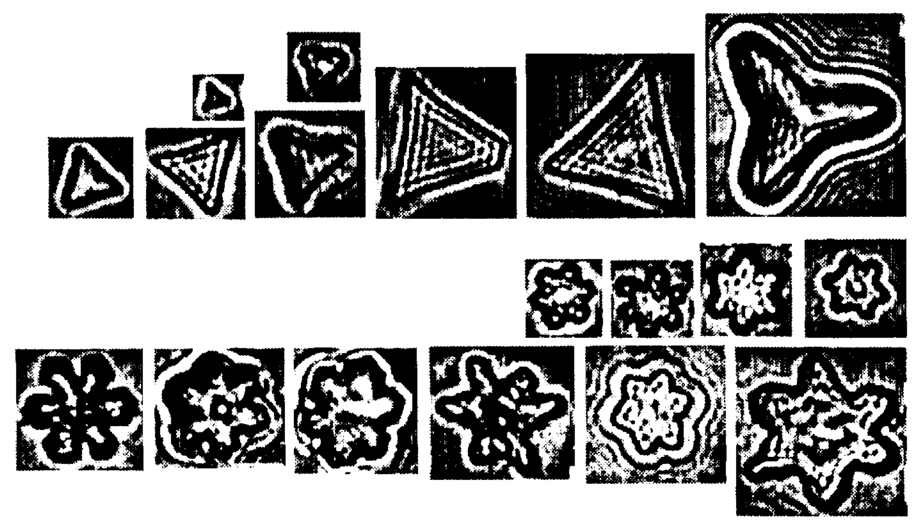


Figure 3. Replica of ice crystals from an evaporating contrail showing trigonal symmetry of plate ice crystals.

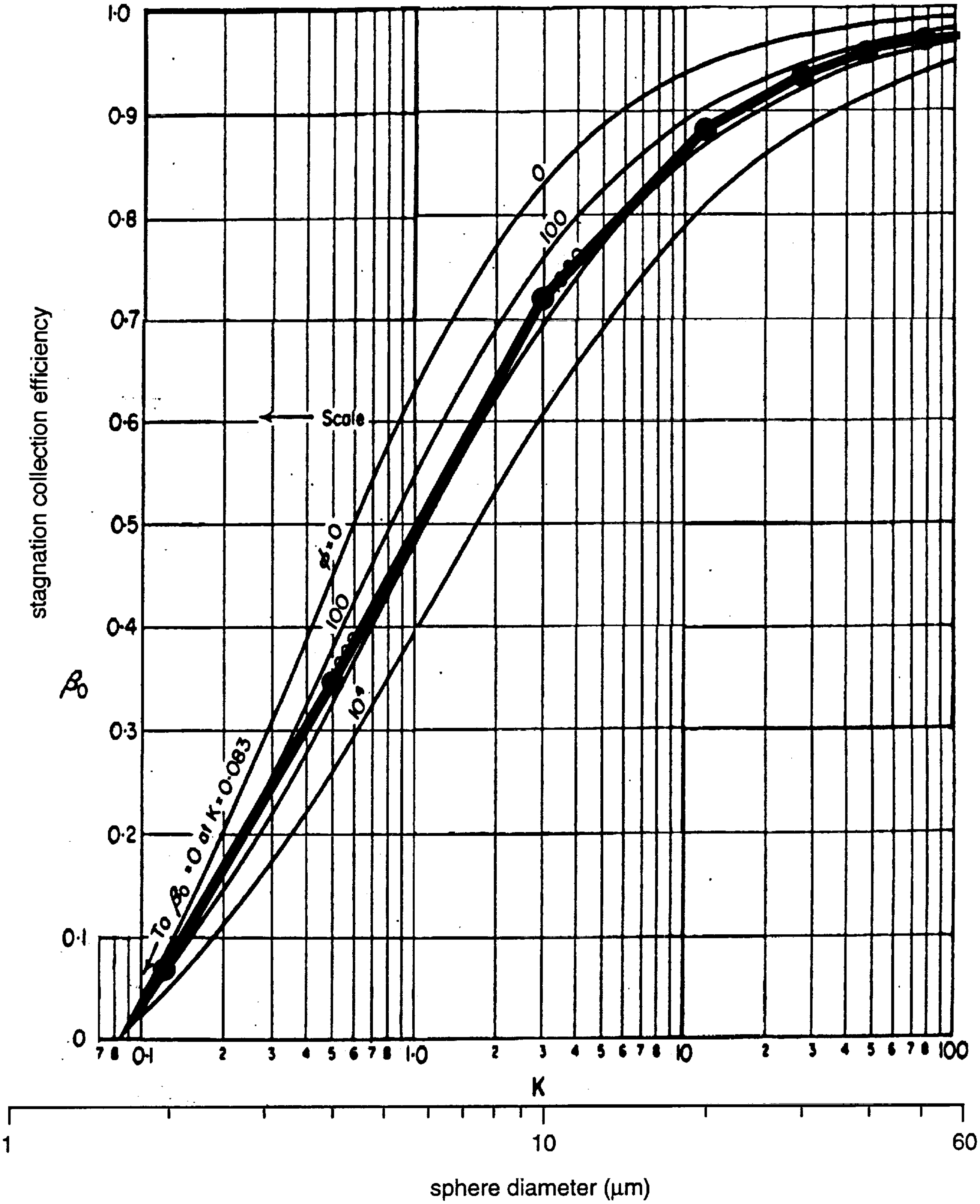


Figure 5a. Computed collection efficiency for spheres of density 0.9 g/cc for collection on cloudscope at air speed of 200 m/s.

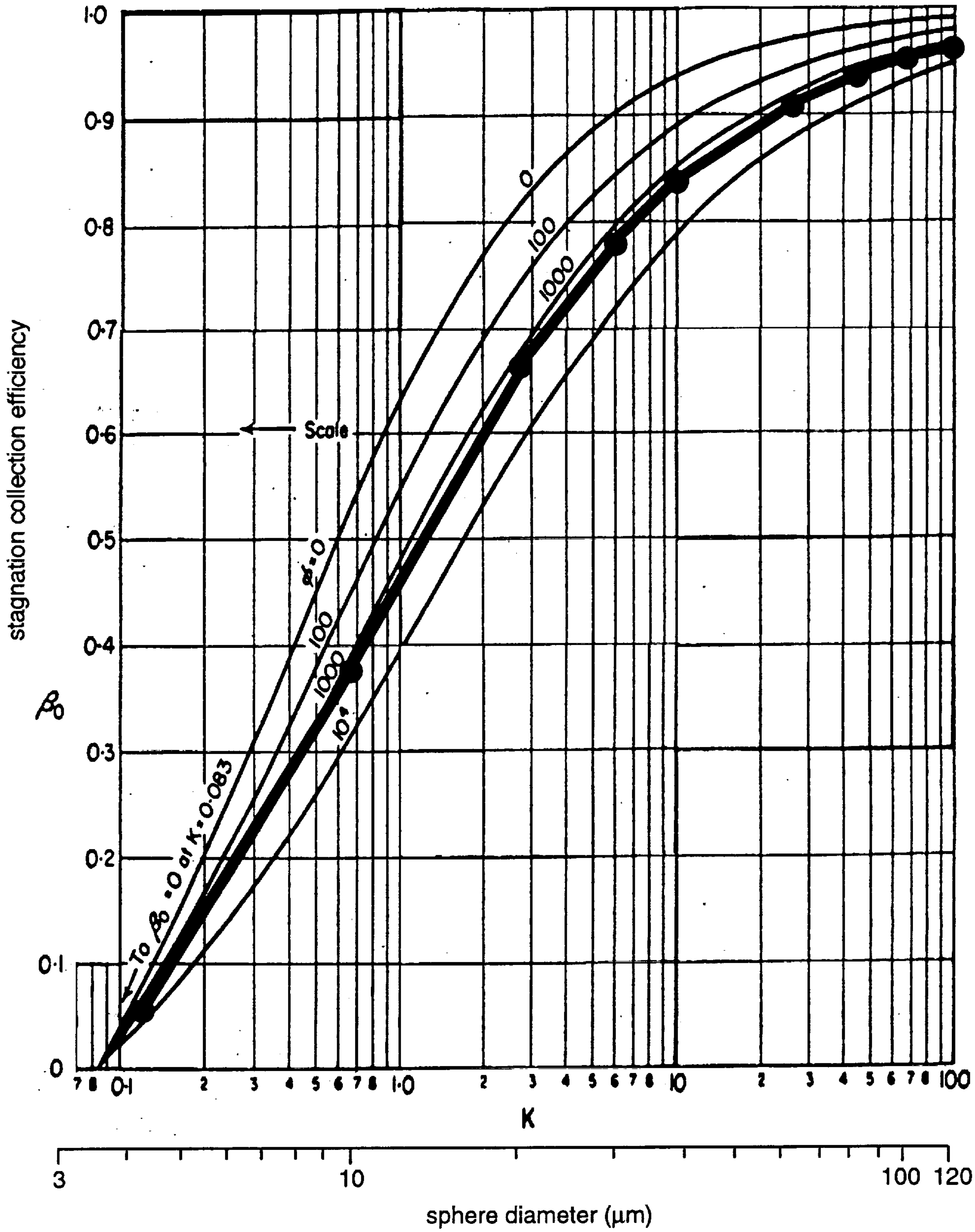


Figure 5b. Computed collection efficiency for spheres of density 0.2 g/cc for collection on cloudscope at air speed of 200 m/s.

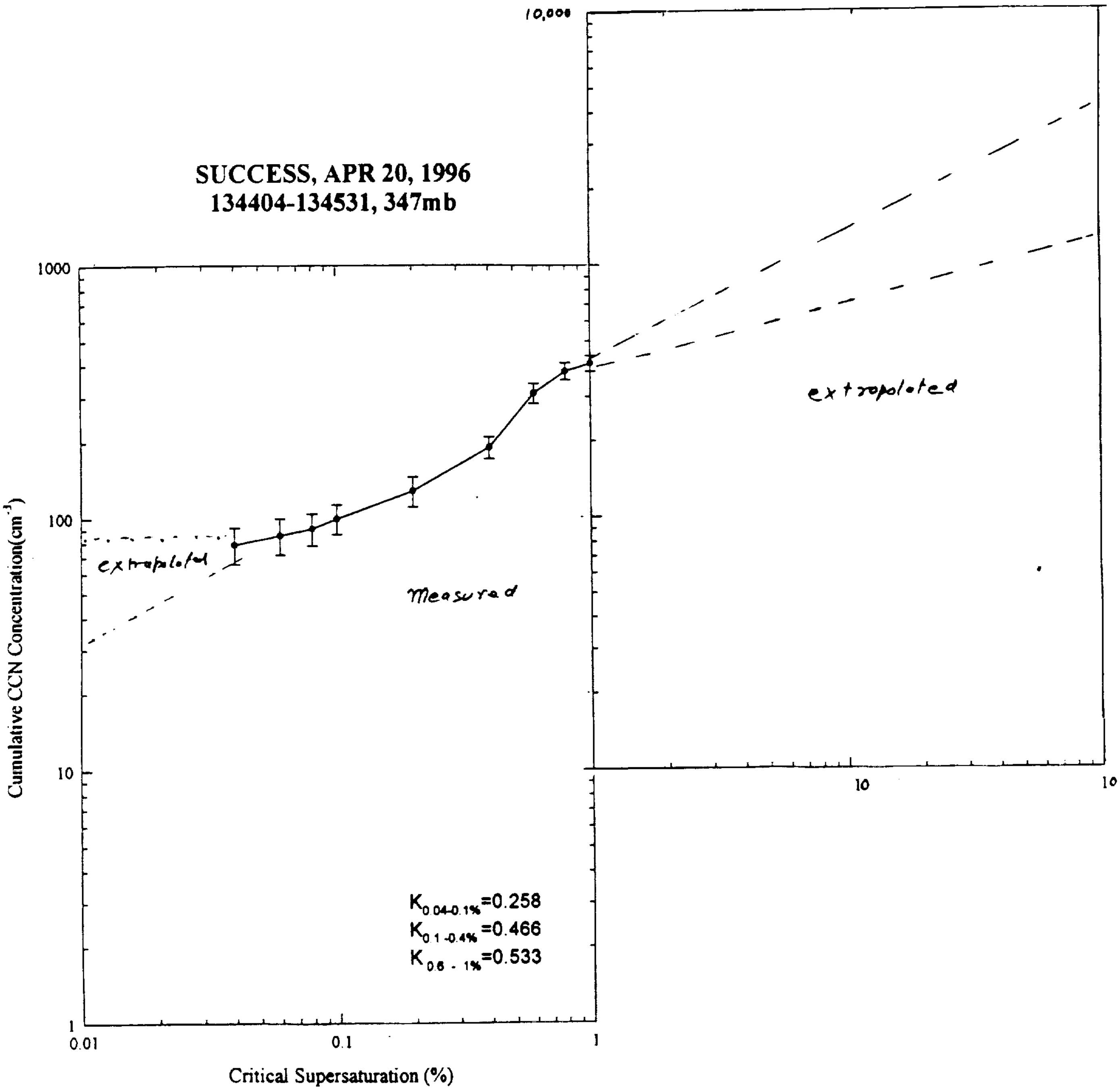


Figure 6. CCN spectra from an old contrail penetration.
April 20, 1996: 13 44 04 - 13 45 31, 347 mb, -58°C.
The data are extrapolated to higher supersaturation as illustration of the possible range of results.

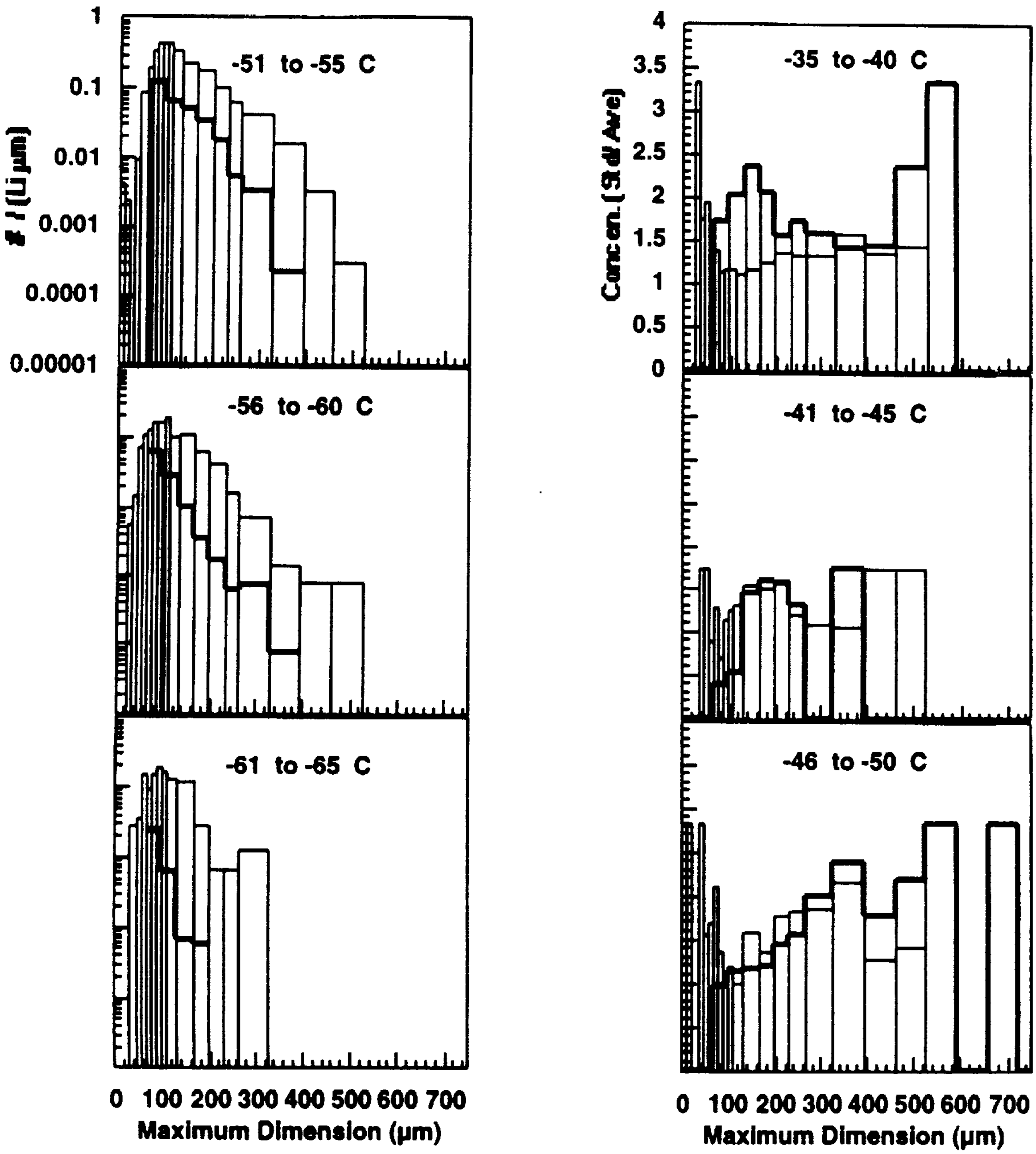


Figure 7. Size distribution standard deviation normalized by the average distributions for replicator and 2DC (thick lines), for the indicated temperature ranges.

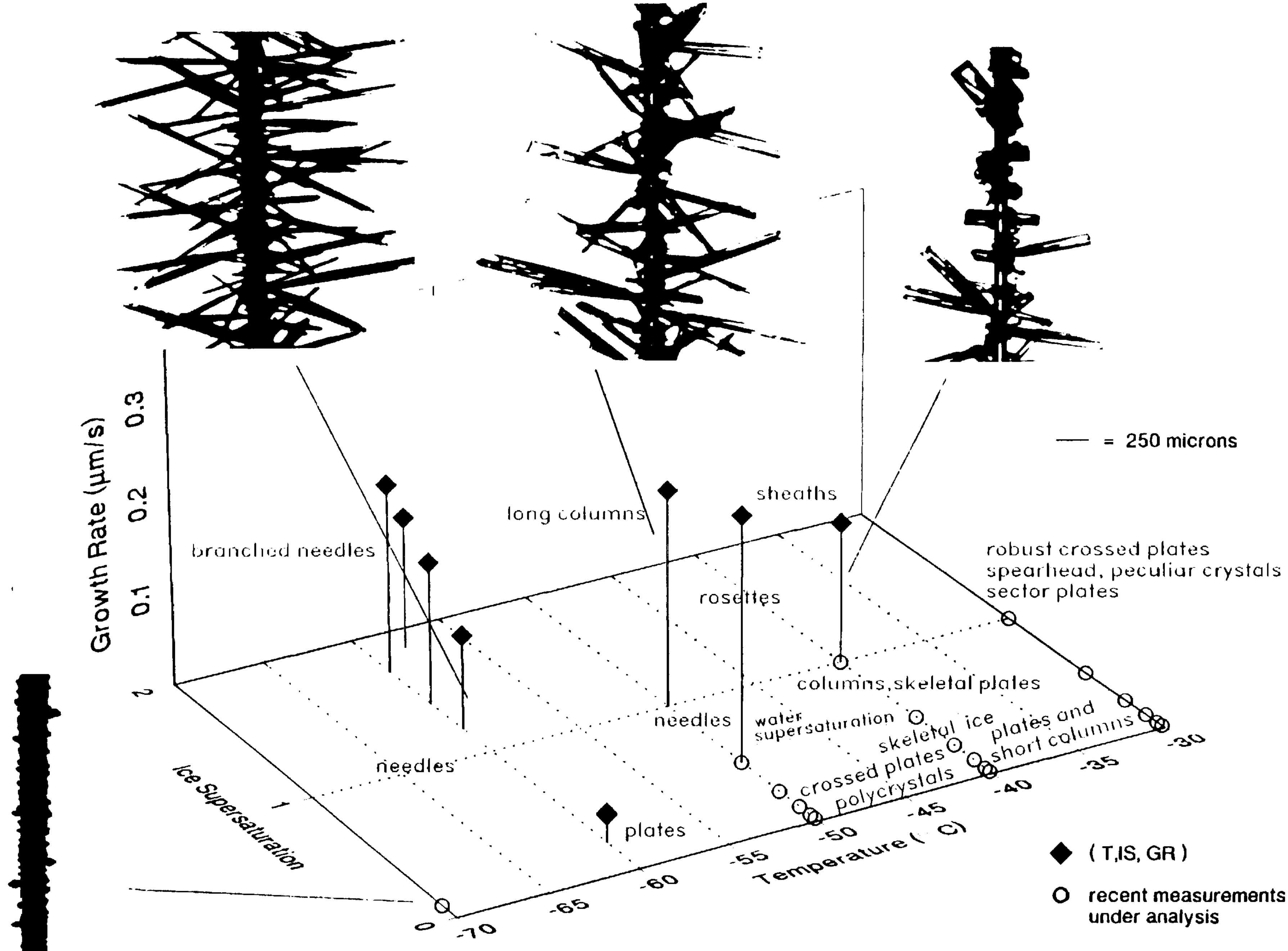


Figure 8.

Summary of the ice crystal habits and growth rates for -30°C to -70°C and a pressure of 100 mb.

IX **Table 1**

SUCCESS REPLICATOR/CLOUDSCOPE/CCN Data Log April/May 1996 DC-8
JH/JHu Mar 97; rev 8

Flt #	date	day	Repl Video	Repl	CS Vid	C CN	Data Tape	Mission
960201	Apr 10	W	0	0	2	0	(1)	Ames Or,Wa
02	Apr 13	Sa	1	*	0	*	1	Ferry to Salina
03	Apr 15	M	1	*	0	*	1	clr sky Coord.
04	Apr 16	Tu	1	*	0	*	1	Thin Cirrus,T39
05	Apr 18	Th	1	*	2	*	1	Cirrus unicus OK,TX
06	Apr 20	Sa	1	*	3	*	1	Cirrus CART
07	Apr 21	Su	0	0	3	*	2	Conv in outfl TX OK
08	Apr24	W	1	*	4	*	1	Hi cirrus OK CART
09	Apr 27	Sa	0	0	3	*	1	Thick cirrus OK CART
10	Apr 29	M	0	0	0	*	2	Lo stratus OK CART
11	Apr 30	Tu	1	*	2	*	2	757 ex/ Wave NM
12	May 2	Th	3	*	2(3)	*	1	Wave Cl CO
13	May 3	F	2	*	2	*	1	757 OK CART
14	May 4	Sa	2	*	2	0	1	757 OK CART
15	May 7	Tu	0	0	2	0	(1)	757 NE
16	May 8	W	0	0	2	*	1	Conv outfl;Contr.10 WI
17	May10	F	2	*	2	*	1	Transit to Ames
18	May12	Su	0	0	3	*	1	Self contr OR;Ocean
19	May15	W	0	0	3	*	(1)	OR, WA Cirrus

Key:

C Cloud Condensation Nucleus continuous measurements between
CN 0.02% and 1% supersaturation, 35 channels, time resolution 3 s.

Repl: Formvar replicator: approx 2 hours of data; video tape gives timing.

CS: Cloudscope: each tape 2 or 3 hours.

* Data available.

0 No data
Instrument not flown or inoperative.
Tapes not available at DRI (n), n = number of tapes.

Nov 97 rev8

SUCCESS flt # 9602xx	date 1996	CS tape	times, Z	CS Offset from DADS instrument position
01	10 April	1/2	21 40 45 - 23 22 53	time 11s behind DADS
		2/2	23 24 08 - 01 04 53 (11 Apr)	R I
05	18 April	1/2	17 06 46 - 19 10 15	?
		2/2	19 11 09 - 20 30 18 (data image follows)	R I
06	20 April	1/3	15 33 46 - 17 19 45	time 24 s ahead of DADS
		2/3	17 20 30 - 19 30 42	
		3/3	19 14 50 - 20 56 34	
07	21 April	1/3	17 34 35 - 19 37 24	time 7s behind DADS
		2/3	19 39 25 - 21 42 49	R I
		3/3	22 45 10 - 23 54 49	
08	24 April	1/4	16 45 20 - 18 43 11	time 8s ahead of DADS
		2/4	18 44 12 - 20 47 28	R I
		3/4	20 49 46 - 22 49 40	
		4/4	22 50 11 - 23 56 34	
09	27 April	1/3	16 07 26 - 18 13 24	time 17s ahead of DADS
		2/3	18 14 00 - 20 17 29	R I
		3/3	20 18 55 - 22 14 19	
11	30 April	1/2	17 19 50 - 19 23 14	time 1s ahead of DADS
		2/2	19 34 26 - 22 35 45	R I
12	2 May	1/3	16 51 20 - 18 21 49	time 5 s ahead of DADS
		2/3	not available; only 19 39 50 to 19 39 56	R I
		3/3	20 29 31 - 22 21 38	
13	3 May	1/2	17 25 51 - 20 29 02	time 10s ahead of DADS
		2/2	20 31 19 - 22 29 36	R I
14	4 May	1/2	17 10 14 - 20 26 51	time 13 s ahead of DADS
		2/2	20 28 05 - 21 39 46	R I
15	7 May	1/2	18 16 35 - 21 17 59	time 27 s ahead of DADS
		2/2	21 18 59 - 23 21 56	R I

16	8 May	1/2	17 15 16 - 19 18 38	time 27 s ahead of DADS
		2/2	19 20 52 - 21 15 22	R I
17	10 May	1/2	00 10 05 - 03 12 32	time 31 s ahead of DADS
		2/2	15 13 16 - 16 51 30	R I
18	12 May	1/3	20 37 41 - 22 42 08	time as DADS
		2/3	22 43 06 - 00 46 34 (13 May)	R I
		3/3	00 47 55 - 01 46 26 (13 May)	
19	15 May	1/3	19 18 27 - 21 13 57	time as DADS
		2/3	21 14 38 - 23 15 26	R I
		3/3	23 16 32 - 00 34 33 (16 May)	

DC-8 INSTRUMENT POSITION (top view):

L O: Left Outboard; L O: Left Outboard.
R I: Right Inboard; R I: Right inboard.

```
              X
              X
           X  X  X
         X    X     X
      X       X       X
 L O   L I    X     R I   RO
              X
              X
           XXXXX
```

Flt #	date	LO	LI	RI	RO
960201	Apr 10	CS	VIPS	REPL	MASP
960202	Apr 13	?	?	REPL	MASP
960203	Apr 15	2DC	VIPS	REPL	MASP
960204	Apr 16	2DC	VIPS	REPL	MASP
960205	Apr 18	2DC	REPL	CS	MASP
960206	Apr 20	2DC	REPL	CS	MASP
960207	Apr 21	2DC	VIPS	CS	MASP
960208	Apr 23	2DC	REPL	CS	MASP
960209	Apr 27	2DC	VIPS	CS	MASP
960210	Apr 29	?	?	VIPS	?
960211	Apr 30	REPL	VIPS	CS	MASP
960212	May 2	REPL	VIPS	CS	MASP
960213	May 3	REPL	VIPS	CS	MASP
14	May 4	REPL	FSSP	CS	MASP
15	May 7				
16	May 8				
17	May 10				
18	May 12	2DC	VIPS	CS	MASP

Detail of possible replica times:
(lo speed = 0.5 cm s^{-1}; hi speed = 1 cm s^{-1}; approx 70m film per flt.)

960202 18 54/19 00 lo; 19 00/19 55 hi; 19 55/20 20 lo
Ap 13

960204 17 18/18 41 hi; 18 41/19 36 lo; 19 36/19 48 hi.
Ap 16

960205 18 00/18 25 hi; 18 25/19 40 lo; 1940/19 50 hi; 19 50/19 52 lo
Ap 18 19 52/19 54 hi; 19 54/20 01 lo; 20 01/20 15 hi; 20 15/20 37 lo

960206 16 55/17 19 lo; 17 19/17 23 hi;17 23/17 33 lo;
AP 20 17 33/19 05 hi;19 05/19 25 lo.

190208 18 55/19 01 lo; 19 01/19 56 hi; 19 56/20 23 lo.
Ap 24

190211 questionable data
Ap 30

190212 17 10/17 37 lo; 17 58/18 07 lo; 18 10/18 20 lo;18 30/18 40 lo;
May 2 19 03/21 40 lo; with 19 40, 20 18, 20 20, 20 30, 21 18, 2 min hi

190213 18 32/ 22 30 lo.
May 3

960214 17 50/21 44 lo
May 4